

Deterministic growth model of Laplacian charged particle aggregates

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(Received February 7, 2008)

The results of the computer simulation of the aggregates growth of the similarly charged particles in the framework of deterministic Laplacian growth model on a square lattice are presented. Cluster growth is controlled by three parameters p, E, λ , where p - Laplacian growth parameter, E - energy of a particle sticking to a cluster, λ - the screening length of electrostatic interactions. The phase diagram of cluster growth is built in the co-ordinates E, λ . The zones of different cluster morphology are selected: I-the zone of finite X-like structures, II-the zone of infinite ramified structures, controlled by electrostatic interactions, III-the zone of infinite structures with electrostatic interactions effectively switched off. Simple electrostatic estimations of the locations of the zone boundaries are presented. It is shown that in general case within the zone II the continuous change of D_f , controlled by parameters p, E, λ , takes place. In the degeneration limit when the given model transforms into deterministic version of the Eden model (at $p = 0$), the crossover from linear ($D_f = 1$) to compact ($D_f = 2$) structures is observed when passing through the boundary between the zones I and II.

PACS number(s): 02.70.Lq, 68.70.+w, 75.40M

A number of models have been developed to describe the growth of aggregates with complex, ramified structure. The most important among them are the models of diffusion-limited aggregation fractal growth (DLA model), compact growth (Eden model), ballistic growth, dendrite, and dense-branching structure growth [1]. On changing the local conditions controlling the process of particle's sticking to a cluster, it is possible to observe the structures with different morphologies [2–4]. The study of aggregation, which takes into account the specific interactions between the particles, e.g., the interactions of electric or magnetic origin, is of a special interest. Such interactions may effectively manifest themselves in colloids [5–7], in electrochemical deposition phenomena [4], in discharge phenomena (in particular, in ball lightning development from unipolar plasma [8]).

This work presents the results of the growth study of aggregates formed by the likely charged particles within the framework of deterministic Laplacian growth model. Such model is widely used to describe the growth of dendrite-like structures [9,10]. The model of such type allows to obtain the deterministic analogue of DLA-type fractal patterns. The choice of deterministic model allows also to enhance the lattice-imposed anisotropy of the patterns and to suppress the noise effects.

Our model takes into account the screening effects by limiting the radius of electrostatic repulsion between the particles as $r < \lambda$. Each simulation step included the numerical solving of Laplace equation on a lattice and the calculation of sticking probability f (\propto to the normalized gradient of the potential) all over the cluster perimeter. Before the sticking of a new particle to the point (j) of a cluster, the calculation of electrostatic repulsion energy $U(j)$ between the new particle and all the other parti-

cles of cluster formed on the previous evolution step was carried out:

$$U(j) = \sum_i q^2 a_{ij} / r_{ij}, \quad (1)$$

where q is a particle charge, r_{ij} is a distance between the i particle of the aggregate and the j -point, $a_{ij} = 1$ at $r_{ij} = \lambda$ and $a_{ij} = 0$ otherwise.

New particle joined the cluster at the (j)-point of perimeter only when two conditions were satisfied: a) $f > p$ in this point, and b) $U(j)$ did not exceed some critical value E . Here, p is the Laplacian growth parameter, $0 < p < 1$. Note that E corresponds to the short-range energy of single particle attraction to a cluster, and that this attraction may result from the Van der Waals interactions.

The growth of clusters took place on the two-dimensional square lattice. The distances were measured in the lattice units, and all the particles had a charge q equal to 1. Thus, we may consider that λ, E and U parameters introduced above are dimensionless. The lattice size comprised 1000×1000 and the number of cluster particles did not exceed $5 \cdot 10^5$.

So, in this model the three parameters, λ, E, p control the growth of clusters. In the regular deterministic Laplacian growth model, assuming no electrostatics, the growth of clusters is controlled only by the value of parameter p and their size is not limited. At $p = 0$, our model corresponds to deterministic Eden model and gives the compact clusters with fractal dimensionality $D_{fo} = 2$. At $0 < p < 1$, the structures with fractal dimensionality develop; their dimensionality decreases with the increase of p [9], [10]. For $p = 0.3$, which case will be analyzed in more details below as an example, we have

obtained, $D_{fo} = 1.49$.

The switch-on of electrostatic interactions results in considerable complication of the growth patterns. In our case, both finite and infinite clusters may form, depending on the values of λ, E, p parameters.

Figure 1 presents the phase diagram in $\lambda - E$ coordinates, where the zones of different cluster morphology are distinguished. The boundary between the zones II and III is shown by the line 1 for $p = 0.3$ and by the line 1' for $p = 0$. The cluster patterns from the different zones of diagram (marked by the symbol ■ and relevant Latin characters) are presented on Fig.2. All clusters have specific anisotropic square-like shape due to deterministic version of a growth model for clusters growing on a square lattice and in a given presentation of clusters the lattice axes are directed along the square diagonals.

In the zone I, the growth of finite X-like structures is observed, and in the zones II and III the growth of infinite clusters takes place. The maximal cluster size limitation observed in the zone I may be explained as follows: at high enough λ values (or, correspondingly, small enough E) the electrostatic repulsion energy at the points of new particles sticking U continuously grows with cluster growth. At some critical value of cluster size the realization of condition $U > E$ occurs at all the sites of cluster perimeter, which means the halt of the cluster growth. The development of non-ramified X-like structures in the zone I may be explained by almost complete control of the cluster growth by electrostatic repulsion forces in the given area of λ, E values. Under such conditions, the particles stick to the cluster predominantly at points most distant from the cluster center, where the energy of electrostatic repulsion is the lowest. In this case, the growth of clusters proceeds mainly along the lattice axes, i.e., on the lines directed along the diagonals of the square.

At $\lambda = \infty$, only the growth of finite clusters with the size controlled by parameters E and p takes place, and the ramification of structures is also possible. The dash-filled area on Fig.1 approximately corresponds to the zone of transition to the areas of infinite cluster growth. Note that the location of this zone practically does not depend on the value of the parameter p .

The zone of infinite clusters, where fractal, dense-branching and dense structures develop, should be divided into two areas - II and III. The necessity of such division follows from the detailed analysis of fractal and morphological properties of clusters in these areas.

The estimation of the fractal dimensionality D_f of the deterministic growth patterns requires some caution. We used the "sand-box" method for D_f estimation and analyzed the number of particles in the squares of ascending size [11]. Application of the squares instead of circles commonly used in this method allows to consider more precisely the effects of cluster anisotropy imposed by the symmetry of square lattice. Besides, as it is seen from Fig.2, the pattern structures may be subdivided quite clearly, especially at large E values, into the dense X-

like nucleus (with $D_f = 2.0$) and ramified peripheral part. We calculated the fractal dimensionality only for ramified part of a pattern.

Figure 3 presents dependencies of D_f on E for $\lambda = 10, 100$ (curves 1 and 2) and D_f on λ for $E = 100$ (curve 3) in the case of $p = 0.3$. When moving across the phase diagram (Fig.1) along the E axis at some constant value of λ (same as at $\lambda = \infty$), the clusters with fractal dimensionality $D_f = 1$ are observed in the zone I. The D_f sharply increases on passing into the zone II; further on, the decreasing of D_f takes place and at some point $E = E_{II-III}$, which corresponds to the point of transition into zone III, the fractal dimensionality reaches the value $D_{fo} = 1.49$. As it was mentioned above, this value corresponds to the fractal dimensionality of clusters that grow at $p = 0.3$ without electrostatic interactions. When moving along the λ axis at some fixed value of E , the clusters characterized by the constant value of fractal dimensionality, $D_{fo} = 1.49$ are found in the zone III. But at some point $\lambda = \lambda_{II-III}$, which corresponds to the point of transition into the zone II, a rather sharp increase of D_{fo} takes place.

The similar behavior of the fractal dimensionality is observed for other non-zero values of p , though localization of the boundary between the zones II and III depends on the p value. The D_f behavior somewhat differs at $p = 0$. In this case, the clusters of the zone I are characterized by $D_f = 1$ (same as for $\lambda = \infty$), but $D_f = 2$ in the zones II and III. Thus, on passing from the zone I into the zone II across the dash-filled zone, a crossover between the linear ($D_f = 1$) and dense structures ($D_f = 2$) is observed. The clusters of the zone II have a dense-branching morphology and their structure becomes more compact as far as the system approaches the boundary of the zone III (the curve 1' on the Fig.1). Zone III is characterized by the growth of compact clusters of the square shape only, which is practically equivalent to the cluster growth conditions with switched-off electrostatic interactions.

As it follows from the above discussion, zone II of the phase diagram corresponds to the strong effect of electrostatic interactions on a cluster growth, whereas zone III is the zone of cluster growth under completely switched-off electrostatic interactions. Here, we propose the simple electrostatic estimations for explanation of the observed behavior.

The absence of any considerable effect of electrostatic interactions on the growth of clusters inside the zone III may occur only if the points of cluster-growth halt, complying with the condition of $U > E$, are absent on the cluster perimeter. Then, the boundary between the zones II and III may be estimated out of the condition $E_{II-III} < U$. The highest electrostatic repulsion is peculiar to perimeter points coinciding with the middle points of the square sides (Fig.2), since these perimeter points are the nearest to the cluster center. To calculate the electrostatic repulsion energy U in these points, we pass from summation to integration in relation (1) and assume

the ideal density profile for fractal clusters on receding from the center

$$\rho(R) \approx R^{D_f-2}, \quad (2)$$

where R is the distance from the cluster center to the middle-point of the cluster side.

The calculations show that

$$E_{II-III} \approx U = \pi \lambda^{D_f-1} f(D_f), \quad (3)$$

where $f(D_f)$ is the function of cluster fractal dimensionality, D_f . Figure 4 demonstrates the form of this function $f(D_f)$, obtained through the numerical integration.

Equation (3) defines the theoretical boundary between the zones II and III. This boundary is shown on the Fig.1 for the case of $p = 0.3$ by the dashed line 2. We see that this theoretical boundary line goes above the real boundary shown by the line 1, which is not surprising since the density profile assumption (2) does not take into account the existence of the compact nucleus.

On $p = 0$, i.e., for the case of deterministic analogue of the Eden model, the growth of compact clusters is observed, and we obtain from (3)

$$E_{II-III} \approx U = \pi \lambda. \quad (4)$$

Condition (4) defines the boundary between the zones of compact clusters growth and dense-branching morphology. In this case, the theoretical boundary defined by (4) perfectly describes the simulation data (line 1').

Authors thank Dr. N. S. Pivovarova for valuable discussions of the manuscript and help with its preparation.

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FIG. 1. Phase diagram of Laplacian charged particle aggregates for deterministic growth model presented in the following co-ordinates: screening length, λ against sticking energy, E . The zones of different pattern growth morphology are distinguished: I is the zone of finite X-like structures, II is the zone of infinite ramified structures with cluster growth controlled by electrostatic interactions, III is the zone of infinite structures with effectively switched-off electrostatic interactions. Symbol ■ marks the points corresponding to the clusters presented on the Fig. 2, which were grown under the following conditions: $\lambda = 100 - E = 14(a), 40(b), 100(c)$; $\lambda = \infty - E = 50(a'), 100(b'), 500(c')$. The solid lines 1 and 1' show the boundaries between the zones II and III for $p=0.3$ and $p=0$, correspondingly. The dashed line 2 shows the theoretically predicted position of this boundary for $p=0.3$.

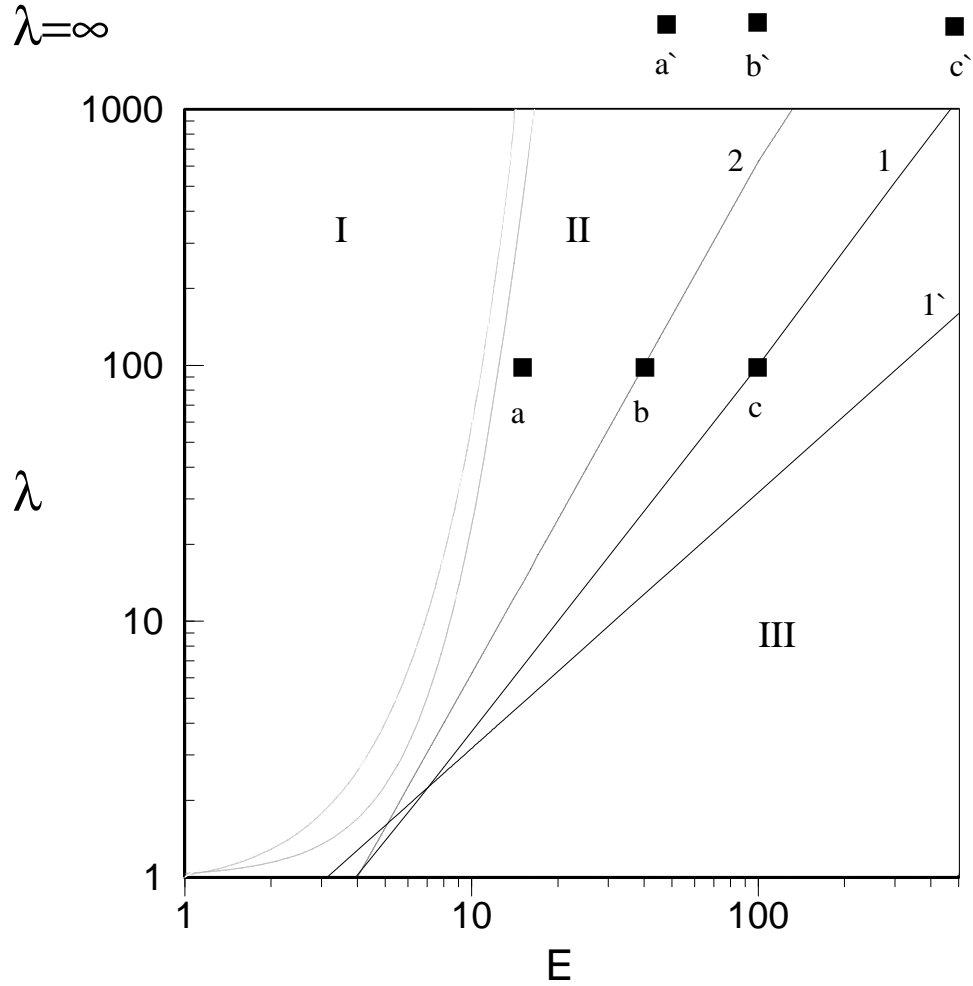
FIG. 2. Typical cluster patterns for the model of deterministic Laplacian growth of charged particles aggregates obtained at different values of p, E, λ (which are shown on the figure). Characters a, a', b, b', c and c' correspond to the points, shown on the phase diagram (Fig.1).

FIG. 3. Dependencies of the fractal dimensionality D_f versus the sticking energy E at the fixed values of $\lambda = 10(1), 100(2)$ and versus the screening length λ at the fixed value of $E=10$ (3).

FIG. 4. Profile of $f(D_f)$ from the equation (3). This function is used for theoretical estimation of the boundary between the zones II and III on the phase diagram (Fig.1).

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$\lambda=\infty$



$p=0.3$

$p=0$

$\lambda=100$

$\lambda=\infty$

$\lambda=\infty$

